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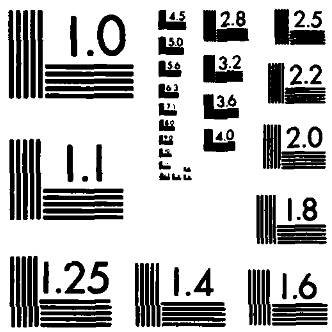

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AD-A135 687

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THE OPTICAL SPECTRA OF AEROSOLS

Pricipal investigator: F. Borghese

Contractor: F. Borghese

Contract n° DAJA37-81-C-0895

Fifth Periodic Report

March 1983 – October 1983

The research reported in this document has been made possible through support and sponsorship of the US Government through its European Research Office of the US Army. This report is intended for internal management use of the contractor and of the US Government.

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R&D 4103-EN

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER	2. GOVT ACCESSION NO. A135687	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) The Optical Spectra of Aerosols		5. TYPE OF REPORT & PERIOD COVERED Interim
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) Prof. F. Borghese		8. CONTRACT OR GRANT NUMBER(s) DAJA37-81-C-0895
9. PERFORMING ORGANIZATION NAME AND ADDRESS Istituto di Struttura della Materia Universita di Messina Messina, Italy		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 61102A-IT161102-BH57-01
11. CONTROLLING OFFICE NAME AND ADDRESS USARDSG-UK Box 65 FPO NY 09510		12. REPORT DATE Oct 83
		13. NUMBER OF PAGES 4
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)		15. SECURITY CLASS. (of this report) UNCLASSIFIED
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited.		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Aerosols, optical scattering, optical spectra, optical constants		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) A cluster of spheres is a model scatterer suitable to approximate the properties of a nonspherical particle. An assembly of such spheres should therefore be a good approximation of an actual aerosol composed of nonspherical particles. Details of the calculation process are included in an annex entitled "Macroscopic optical constants of a cloud of randomly oriented nonspherical scatterers."		

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In the last six months we were able to complete our work on the calculation of the macroscopic optical constants of an assembly of clusters of random orientation. As mentioned in the preceding Reports, a cluster of spheres is a model scatterer suitable to approximate the properties of a nonspherical particle. An assembly of such clusters should therefore be a good approximation of an actual aerosol composed of nonspherical particles. The work done in the last semester has mainly required the translation of the formulas reported in the paper attached to the preceding Report into a set of computer programs. As this has been done, the theory has been applied to a few model aerosols chosen so as to yield a general in sight in to the kind of the results obtainable through our approach. A careful through analysis of the results supports the reliability both of the theory and of the computer programs. The details of the calculations are reported in the enclosed paper(1) which has been submitted to "Il Nuovo Cimento". Our results have also been presented at the 69.th Annual Meeting of the Italian Physical Society(2). In the subsequent discussion it has been suggested that our approach could be applied to the study of the absorption from clouds of interstellar dust, an optical medium whose physical properties meet the requirements for the full applicability of the theory.

We want to enfasize that the programs we used to get the above mentioned results do not include the use of the symmetry properties of the clusters. Therefore our next step shall be the inclusion of the symmetry into our programs, in order to allow us to study aerosols composed of relatively big clusters. The above mentioned calculations are, indeed, the most complicated that can be done without the help of group theory. Furtermore, we notice that the approach we used to connect the scattering properties of a single cluster to the macroscopic properties of the aerosol is quite elementary. A

possible line of development and improvement of our work could be the use of a more sofisticated approach for the connection of microscopic and macroscopic properties.

REFERENCES

1) F.Borghese, P.Denti, R.Saija, G.Toscano and O.I.Sindoni: Costanti ottiche di un aerosol molecolare; Bollettino SIF 130, 20th September 1983.

2) F.Borghese, P.Denti, R.Saija, G.Toscano and O.I.Sindoni: Macroscopic optical constants of a cloud of randomly oriented nonspherical scatterers; submitted to "Il Nuovo Cimento".

Annex

Received after the Fourth Periodic Report	$ 2500
Publication of the paper of ref. (2)	1200
Computer time	2000
Remainder	$ 300

PACS 42.20

MACROSCOPIC OPTICAL CONSTANTS OF A CLOUD OF RANDOMLY ORIENTED NONSPHERICAL SCATTERERS(*)

by

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Summary.

A method to calculate the macroscopic optical constants of a low density medium consisting of a cloud of identical nonspherical scatterers is presented. The scatterers in the medium are clusters of dielectric spheres and the electromagnetic field scattered by each of the clusters is obtained as a superposition of multipole fields, as previously proposed by the authors. The transformation properties of the spherical multipoles under rotation allow the orientational-dependent terms in the expression for the forward-scattering amplitude of each of the clusters to be factored out. In this way the sum of the scattering amplitudes of the clusters with different orientations, needed to calculate the optical response of the medium,

is greatly facilitated and admits a simple analytic expression in the case of randomly oriented clusters. Results of calculations of the optical constants for a few model media are presented.

(*) Based on work supported in part by the CNR through the GNSM and in part by the U.S. Army European Research Office through contract DAJA37-81-C-0895.

1. INTRODUCTION

It is well known that the key quantity for the evaluation of the optical properties of a random distribution of scatterers constituting a low density medium , such as e.g. an aerosol, is the electromagnetic scattering amplitude matrix of the individual scatterers. In fact, the relevant optical parameters of such a medium, namely the refractive index and the absorption coefficient, can be in principle obtained from the sum of the forward-scattering amplitudes of each of the scatterers[1,2]. When these have spherical symmetry their scattering amplitudes are determined from the Mie theory[1,2,3] and the sum leading to the optical parameters presents no additional difficulty on account of the assumed low density of the distribution. The Mie theory, however, does not seem satisfactorily applicable to real aerosols, for the experimental data support the conclusion that the particles of the most common aerosols behave as nonspherical scatteres[4,5].

In this paper we introduce a method to calculate the relevant optical constants of a medium consisting of a low density cloud of identical nonspherical scatterers in the case that each of them is a cluster of dielectric spheres. These model scatterers proved to be computationally sound and to approximate some kind of nonspherical scattering centers:

e.g. they hold a dependence of their total cross section on the direction of the propagation vector of the incident field.[6,7,8,9] Moreover the expression of the scattering amplitude of each cluster can be given in terms of the amplitudes of the multipole fields scattered by the cluster itself and does not require too much computational effort. The literature does not report calculations of the optical behaviour of a medium of nonspherical scatterers, but in the case they are all oriented alike[10]. Indeed, identical nonspherical scatterers have a different forward-scattering amplitude depending on their orientation. This greatly complicates the sum over the scattering amplitudes needed to calculate the optical response of a cloud of such objects[1]. The medium we proposed above allows us to overcome these difficulties and to calculate its optical constants even when the clusters have different orientations.

Therefore, in section 2, we recall the theory to calculate the amplitudes of the multipole fields scattered by a single cluster[6,7] and put the final equations in a form suitable for our purposes. In section 3, we exploit the transformation properties of the multipole fields under rotation[11] and succeed in factoring out of the expression for the multipolar amplitudes all the terms depending on the orientation of the

cluster. In this way the sum of the scattering amplitudes of clusters of different orientation is greatly facilitated. Furthermore, when both the space distribution and the orientation of the (identical) scatterers are random, the above sum is shown to assume a simple analytic expression. At last, in section4, results of the calculations of the optical constants for a few model media are presented. In particular we devised a simple model medium exhibiting birefringence and dichroism even when its clusters are randomly oriented.

We conclude these introductory remarks observing that no dispersion relation is taken into consideration since our treatment is mainly concerned with the effects of nonsphericity of the scatterers. For the sake of simplicity neither the case of the clusters of conducting spheres is included, on account that such an extension does not present further difficulties or advantages for our purposes.

2. THE FIELD SCATTERED BY A CLUSTER OF SPHERES

In this section we concisely recall how the multipolar amplitudes of the field scattered by a cluster of spheres can

be calculated, referring elsewhere for further details[7]. In the following both the spheres and the matrix including them are assumed to be nonmagnetic, dielectric, homogeneous and isotropic.

We consider a circularly polari a plane wave inciding on the cluster, i.e. the incident fie is

$$\underset{\sim}{E}_\eta^{(i)} = E_o(\underset{\sim}{\varepsilon}_1 + i\eta\underset{\sim}{\varepsilon}_2)\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}} \quad , \tag{2-1a}$$

$$i\underset{\sim}{B}_\eta^{(i)} = \eta\underset{\sim}{E}_\eta^{(i)} \quad , \tag{2-1b}$$

with $\eta = \pm 1$ according to the helicity. For our purposes we need the multipolar expansion of the field of eq.(2-1) around the center of the α-th sphere:

$$\underset{\sim}{E}_\eta^{(i)} = E_o e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\alpha}(\underset{\sim}{\varepsilon}_1 + i\eta\underset{\sim}{\varepsilon}_2)\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{r}_\alpha} =$$

$$= E_o e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\alpha}\sum_{LM} W_{\eta,LM}[j_L(kr_\alpha)\underset{\sim}{X}_{LM}(\underset{\sim}{\hat{r}}_\alpha) + \eta\frac{1}{k}\nabla\times j_L(kr_\alpha)\underset{\sim}{X}_{LM}(\underset{\sim}{\hat{r}}_\alpha)] \quad ,$$

with

$$W_{\eta,LM} = 4\pi i^L(\underset{\sim}{\varepsilon}_1 + i\eta\underset{\sim}{\varepsilon}_2)\cdot\underset{\sim}{X}^*_{LM}(\underset{\sim}{\hat{k}}) \quad . \tag{2-2}$$

In the above equations $\underset{\sim}{R}_\alpha$ is the vector coordinate of the center of the α-th sphere and $\underset{\sim}{r}_\alpha = \underset{\sim}{r} - \underset{\sim}{R}_\alpha$, j_L are the Bessel spherical functions and $\underset{\sim}{X}_{LM}$ are the vector spherical harmonics[2,11,12].

We expand the scattered field as

$$\underset{\sim}{E}_n^{(s)} = \sum_{\alpha LM} [A_{n,\alpha LM} h_L(kr_\alpha) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha) + B_{n,\alpha LM} \frac{1}{k} \nabla \times h_L(kr_\alpha) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha)] \quad , \qquad (2\text{-}3a)$$

$$i\underset{\sim}{B}_n^{(s)} = \sum_{\alpha LM} [B_{n,\alpha LM} h_L(kr_\alpha) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha) + A_{n,\alpha LM} \frac{1}{k} \nabla \times h_L(kr_\alpha) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha)] \quad , \qquad (2\text{-}3b)$$

while within the α-th sphere the field is

$$\underset{\sim}{E}_n^{(\alpha)} = \sum_{LM} [C_{n,\alpha LM} j_L(kn_\alpha r_\alpha) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha) + D_{n,\alpha LM} \frac{1}{n_\alpha^2 k} \nabla \times j_L(kn_\alpha r_\alpha) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha)] \quad ,$$

$$i\underset{\sim}{B}_n^{(\alpha)} = \sum_{LM} [D_{n,\alpha LM} j_L(kn_\alpha r_\alpha) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha) + C_{n,\alpha LM} \frac{1}{k} \nabla \times j_L(kr_\alpha r_\alpha) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}_\alpha)] \quad .$$

In the equations above $A_{n,\alpha LM}$, $B_{n,\alpha LM}$, $C_{n,\alpha LM}$ and $D_{n,\alpha LM}$ are the multipolar amplitudes, h_L are the Hankel spherical functions of the first kind, n_α is the(real,constant) refractive index of the α-th sphere.

An useful addition theorem(13) allows us to expand the field of eqs.(2-3) in the vicinity of the surface of the α-th sphere, then we apply the tangential continuity conditions to $\underset{\sim}{E}$ and $\underset{\sim}{B}$ to determine the multipolar amplitudes. The amplitudes $C_{n,\alpha LM}$ and $D_{n,\alpha LM}$ can be eliminated to obtain the system of linear nonhomogeneous equations

$$\sum_{\alpha'L'M'} [(\delta_{\alpha\alpha'}\delta_{LL'}\delta_{MM'}\mathcal{R}^{-1}_{\alpha'L'} + \mathcal{H}_{\alpha LM;\alpha'L'M'})\, A_{\eta,\alpha'L'M'} + \mathcal{K}_{\alpha LM;\alpha'L'M'} B_{\eta,\alpha'L'M'}] =$$

$$= - E_o e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\alpha} W_{\eta,LM} \quad , \qquad (2-4a)$$

$$\sum_{\alpha'L'M'} [\mathcal{K}_{\alpha LM;\alpha'L'M'} A_{\eta,\alpha'L'M'} + (\delta_{\alpha\alpha'}\delta_{LL'}\delta_{MM'}\mathcal{S}^{-1}_{\alpha'L'} + \mathcal{H}_{\alpha LM;\alpha'L'M'}) B_{\eta,\alpha'L'M'}] =$$

$$= - E_o e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\alpha} n\, W_{\eta,LM} \quad . \qquad (2-4b)$$

In eqs.(2-4)

$$\mathcal{R}_{\alpha L} = \left[\frac{J_{\alpha L}\frac{d}{dr_\alpha}(r_\alpha j_{\alpha L}) - j_{\alpha L}\frac{d}{dr_\alpha}(r_\alpha J_{\alpha L})}{J_{\alpha L}\frac{d}{dr_\alpha}(r_\alpha h_{\alpha L}) - h_{\alpha L}\frac{d}{dr_\alpha}(r_\alpha J_{\alpha L})}\right]_{r_\alpha = b_\alpha} \quad , \qquad (2-5a)$$

$$\mathcal{S}_{\alpha L} = \left[\frac{n_\alpha^2 J_{\alpha L}\frac{d}{dr_\alpha}(r_\alpha j_{\alpha L}) - j_{\alpha L}\frac{d}{dr_\alpha}(r_\alpha J_{\alpha L})}{n_\alpha^2 J_{\alpha L}\frac{d}{dr_\alpha}(r_\alpha h_{\alpha L}) - h_{\alpha L}\frac{d}{dr_\alpha}(r_\alpha J_{\alpha L})}\right]_{r_\alpha = b_\alpha} \quad , \qquad (2-5b)$$

where b_α is the radius of the α-th sphere and $j_{\alpha L}=j_L(kr_\alpha)$; $h_{\alpha L}= =h_L(kr_\alpha)$, $J_{\alpha L}=j_L(kn_\alpha r_\alpha)$.

Furthermore

$$\mathcal{H}_{\alpha LM;\alpha'L'M'} = \sum_\mu C(1,L,L;-\mu,M+\mu)\, G_{\alpha L\, M+\mu;\alpha'L'M'+\mu}\, C(1,L',L';-\mu,M'+\mu), \quad (2-6a)$$

$$\mathcal{K}_{\alpha LM;\alpha' L'M'} = -i\left[\frac{2L+1}{L}\right]^{1/2} \sum_{\mu} C(1, L, L+1; -\mu, M+\mu)\ G_{\alpha L+1\ M+\mu;\alpha' L'\ M'+\mu}$$

$$\times C(1, L', L'; -\mu, M'+\mu) \qquad , \qquad (2\text{-}6b)$$

where

$$G_{\alpha LM;\alpha' L'M'} = (1-\delta_{\alpha\alpha'})\, 4\pi \sum_{\lambda} i^{L-L'-\lambda} I_{\lambda}(LM;L'M')\, h_{\lambda}(kR_{\alpha\alpha'})\ Y^{*}_{\lambda\ M-M'}(\hat{\underset{\sim}{R}}_{\alpha\alpha'}) \quad (2\text{-}7)$$

and the C's are the Clebsch-Gordan coefficients[11,12]. In eq. (2-7) $\underset{\sim}{R}_{\alpha\alpha'} = \underset{\sim}{R}_{\alpha'} - \underset{\sim}{R}_{\alpha}$, and

$$I_{\lambda}(LM, L'M') = \left[\frac{(2L'+1)(2\lambda+1)}{4\pi(2L+1)}\right]^{1/2} C(L', \lambda, L; 0, 0)\ C(L', \lambda, L; M', M-M')$$

are the Gaunt integrals[11,12].

The solutions of the system (2-4) are

$$A_{n,\alpha LM} = E_{o} \sum_{L'M'} \mathcal{J}^{(A)}_{n,\alpha LM;L'M'}\ W_{n,L'M'} \qquad , \qquad (2\text{-}8a)$$

$$B_{n,\alpha LM} = E_{o} \sum_{L'M'} \mathcal{J}^{(B)}_{n,\alpha LM;L'M'}\ W_{n,L'M'} \qquad . \qquad (2\text{-}8b)$$

In the equations above

$$\mathcal{J}^{(A)}_{n,\alpha LM;L'M'} = -\sum_{\alpha'} e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_{\alpha'}} \left(T^{(AA)}_{\alpha LM;\alpha' L'M'} + nT^{(AB)}_{\alpha LM;\alpha' L'M'}\right) \qquad ,$$

$$\mathcal{J}^{(B)}_{n,\alpha LM;L'M'} = -\sum_{\alpha'} e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_{\alpha'}}(T^{(BA)}_{\alpha LM;\alpha' L'M'} + \eta T^{(BB)}_{\alpha LM;\alpha' L'M'}) ,$$

where the matrix

$$\underset{\approx}{T} = \begin{vmatrix} \underset{\approx}{T}^{(AA)} & \underset{\approx}{T}^{(AB)} \\ \underset{\approx}{T}^{(BA)} & \underset{\approx}{T}^{(BB)} \end{vmatrix}$$

is the inverse to the matrix of the system (2-4)

$$\begin{vmatrix} \underset{\approx}{R}^{-1} + \underset{\approx}{H} & \underset{\approx}{K} \\ \underset{\approx}{K} & \underset{\approx}{S}^{-1} + \underset{\approx}{H} \end{vmatrix} ;$$

obviously all the submatrices have the same number of rows. For our purposes we need the multipolar expansion of the scattered field around the origin of the axes. At a distance larger than the overall size of the cluster the addition theorem already used above allows us to write

$$\underset{\sim}{E}^{(s)}_{n} = \sum_{LM}[\mathcal{A}_{n,LM}\, h_L(kr)\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}) + \mathcal{B}_{n,LM}\frac{1}{k}\nabla\times h_L(kr)\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}})] ,$$

$$i\underset{\sim}{B}^{(s)}_{n} = \sum_{LM}[\mathcal{B}_{n,LM}\, h_L(kr)\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}) + \mathcal{A}_{n,LM}\frac{1}{k}\nabla\times h_L(kr)\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}})] ,$$

where

$$\mathcal{A}_{n,LM} = \sum_{\alpha'L'M'} (\mathcal{J}_{0LM;\alpha'L'M'} A_{n,\alpha'L'M'} + \mathcal{L}_{0LM;\alpha'L'M'} B_{n,\alpha'L'M'}) \quad , \qquad (2\text{-}9a)$$

$$\mathcal{B}_{n,LM} = \sum_{\alpha'L'M'} (\mathcal{L}_{0LM;\alpha'L'M'} A_{n\alpha'L'M'} + \mathcal{J}_{0LM;\alpha'L'M'} B_{n\alpha'L'M'}) \quad . \qquad (2\text{-}9b)$$

In eqs.(2-9) the $\mathcal{J}$'s and the $\mathcal{L}$'s are given by eq.(2-6a) and (2-6b), respectively, with j_λ substituted for h_λ and the omission of the factor $(1-\delta_{\alpha\alpha'})$ within the G's of eq.(2-7). With the help of eqs.(2-8), eqs.(2-9) become

$$\mathcal{A}_{n,LM} = E_0 \sum_{L'M'} U^{(A)}_{n,LM;L'M'} W_{n,L'M'} \quad , \qquad (2\text{-}10a)$$

$$\mathcal{B}_{n,LM} = E_0 \sum_{L'M'} U^{(B)}_{n,LM;L'M'} W_{n,L'M'} \quad , \qquad (2\text{-}10b)$$

where

$$U^{(A)}_{n,LM;L'M'} = \sum_{\bar{\alpha}\bar{L}\bar{M}} (\mathcal{J}_{0LM;\bar{\alpha}\bar{L}\bar{M}} \mathcal{T}^{(A)}_{n,\bar{\alpha}\bar{L}\bar{M};L'M'} + \mathcal{L}_{0LM;\bar{\alpha}\bar{L}\bar{M}} \mathcal{T}^{(B)}_{n,\bar{\alpha}\bar{L}\bar{M};L'M'}) , \qquad (2\text{-}11a)$$

$$U^{(B)}_{n,LM;L'M'} = \sum_{\bar{\alpha}\bar{L}\bar{M}} (\mathcal{L}_{0LM;\bar{\alpha}\bar{L}\bar{M}} \mathcal{T}^{(A)}_{n,\bar{\alpha}\bar{L}\bar{M};L'M'} + \mathcal{J}_{0LM;\bar{\alpha}\bar{L}\bar{M}} \mathcal{T}^{(B)}_{n,\bar{\alpha}\bar{L}\bar{M};L'M'}) . \qquad (2\text{-}11b)$$

Concluding this section we specifically enlighten two points. The first remark regards the variables on which the U's and W's in eqs.(2-10) depend. The definition (2-2) show that the W's depend only on $\theta_{\underset{\sim}{k}}$ and $\phi_{\underset{\sim}{k}}$, the polar angles

of $\underset{\sim}{k}$. Tracing through eqs.(2-11) and related definitions, one finds that the U's depend only on the magnitude of $\underset{\sim}{k}$, the n_α's, the b_α's, and on the components of the $\underset{\sim}{R}_\alpha$'s. Then by eqs.(2-10) we succeeded in expanding the multipolar amplitudes as linear combinations of the geometrical factors U with the orientation-dependent coefficients W. The second remark regards the order of the system(2-4). We have $2NL_M(L_M+2)$ equations, where N is the number of the spheres in the cluster and L_M is the maximum L to be retained in the multipolar expansions. Thus the order becomes easily very large and some advices should be applied. For example one could exploit the symmetry of the cluster, as we reported elsewhere[8,9].

3. THE REFRACTIVE INDEX AND THE ABSORPTION COEFFICIENT OF THE MEDIUM

To begin this section we make a remark about the field inciding on each cluster of the medium we are considering: because of the multiple scattering processes this field should also include the fields scattered by all the other clusters. However, disregarding them amounts to a fairly acceptable approximation when the concentration of the scatterers

is sufficiently low[1,2]. Since we deal just with this low density limit, eqs.(2-10) give also the multipolar amplitudes of each cluster belonging to the medium. Nevertheless, nobody could actually calculate the scattering amplitudes of all the clusters, one at time, referred to an unique system of coordinates, and sum them to get the optical response of the medium. This formidable difficulty can be overcome in two steps, however, through the use of the assumed identity of the clusters and of the transformation properties of the multipole fields under rotation.

Fist of all we define a system of coordinates Σ to which the whole medium can be referred. Furthermore, let us refer each cluster to a local set of axes, $\bar{\Sigma}^{(\nu)}$ for the ν-th cluster, chosen so that the local coordinates of the centers of the corresponding spheres in different clusters are identical. We also associate to $\bar{\Sigma}^{(\nu)}$ the system of coordinates $\Sigma^{(\nu)}$ with the same origin, but with the axes oriented as the axes of Σ. Now we can write the electric field incident on and scattered by the ν-th cluster with reference to the system $\bar{\Sigma}^{(\nu)}$ as

$$E^{(i)}_{\eta\nu} = E_o\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu} \sum_{LM} \bar{W}^{(\nu)}_{\eta,LM} [j_L(k\bar{r}^{(\nu)}) \underset{\sim}{X}_{LM}(\hat{\bar{\underset{\sim}{r}}}^{(\nu)}) +$$

$$+ \eta \frac{1}{k} \nabla \times j_L(k\bar{r}^{(\nu)})\, \underset{\sim}{X}_{LM}(\hat{\bar{\underset{\sim}{r}}}^{(\nu)})] \tag{3-1a}$$

and

$$\underset{\sim}{E}^{(s)}_{n\nu} = \sum_{LM} [\bar{\mathcal{A}}^{(\nu)}_{n,LM} h_L(k\bar{r}^{(\nu)}) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{\bar{r}}}^{(\nu)}) + \bar{\mathcal{B}}^{(\nu)}_{n,LM} \frac{1}{k} \nabla \times h_L(k\bar{r}^{(\nu)}) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{\bar{r}}}^{(\nu)})] , \quad (3\text{-}1b)$$

where, according to eqs(2-10), the amplitudes are

$$\bar{\mathcal{A}}^{(\nu)}_{n,LM} = E_o e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu} \sum_{L'M'} \bar{U}^{(A)}_{n,LM;L'M'} \bar{W}^{(\nu)}_{n,L'M'} , \quad (3\text{-}2a)$$

$$\bar{\mathcal{B}}^{(\nu)}_{n,LM} = E_o e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu} \sum_{L'M'} \bar{U}^{(B)}_{n,LM;L'M'} \bar{W}^{(\nu)}_{n,L'M'} . \quad (3\text{-}2b)$$

Since the components of $\underset{\sim}{\bar{R}}^{(\nu)}_{\alpha}$ are independent of ν, neither the $\bar{U}$'s in eqs.(3-2) depend on it; on the contrary the $\bar{W}$'s depend on ν through $\bar{\theta}^{(\nu)}_{\underset{\sim}{k}}$ and $\bar{\phi}^{(\nu)}_{\underset{\sim}{k}}$. The phase factor $e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu}$ refers the amplitude E_o of the plane wave in the origin of Σ to the amplitude in the origin of $\bar{\Sigma}^{(\nu)}$ (of vector coordinate $\underset{\sim}{R}_\nu$). The electric fields can also be written directly with reference to $\Sigma^{(\nu)}$ as

$$\underset{\sim}{E}^{(i)}_{n\nu} = E_o e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu} \sum_{LM} W_{n,LM} [j_L(kr^{(\nu)}) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}^{(\nu)}) +$$

$$+ n \frac{1}{k} \nabla \times j_L(kr^{(\nu)}) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}^{(\nu)})] \quad (3\text{-}3a)$$

and

$$\underset{\sim}{E}^{(s)}_{n\nu} = \sum_{LM} [\mathcal{A}^{(\nu)}_{n,LM} h_L(kr^{(\nu)}) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}^{(\nu)}) + \mathcal{B}^{(\nu)}_{n,LM} \frac{1}{k} \nabla \times h_L(kr^{(\nu)}) \underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}^{(\nu)})] . \quad (3\text{-}3b)$$

It is to be noticed that the W's in eq.(3-3a) do not depend on ν, viz. on the orientation of the cluster; they actually depend only on the polar angles of $\underset{\sim}{k}$ with respect to the axes of $\Sigma^{(\nu)}$, which by definition are parallel to the axes of Σ. Moreover, on account of their transformation properties under rotation, the vector spherical harmonics in eqs.(3-1) and (3-3) can be referred to each other by[11,12]

$$\underset{\sim}{X}_{LM}(\hat{\underset{\sim}{r}}^{(\nu)}) = \sum_{M'} D^{(L)}_{M'M}(\theta_\nu)\ \underset{\sim}{X}_{LM'}(\hat{\underset{\sim}{r}}^{(\nu)}) \qquad , \qquad (3\text{-}4)$$

where the argument θ_ν of the representation matrices of the full rotational group $\underset{\approx}{D}^{(L)}$ represents the Euler angles of the rotation transforming $\bar{\Sigma}^{(\nu)}$ into $\Sigma^{(\nu)}$. Thus, by substituting eq. (3-4) into eqs.(3-1) and comparing with eqs.(3-3), we get

$$W_{n,LM'} = \sum_{M} D^{(L)}_{M'M}(\theta_\nu)\ \overline{W}^{(\nu)}_{n,LM} \qquad ,$$

whose inverse is

$$\overline{W}^{(\nu)}_{n,LM} = \sum_{M'} W_{n,LM'} D^{(L)*}_{M'M}(\theta_\nu) \qquad , \qquad (3\text{-}5)$$

and

$$\mathcal{A}^{(\nu)}_{n,LM'} = \sum_{M} D^{(L)}_{M'M}(\theta_\nu)\ \overline{\mathcal{A}}^{(\nu)}_{n,LM} \qquad , \qquad (3\text{-}6a)$$

$$\mathcal{B}^{(\nu)}_{n,LM'} = \sum_{M} D^{(L)}_{M'M}(\theta_\nu)\ \bar{\mathcal{B}}^{(\nu)}_{n,LM} \qquad . \tag{3-6b}$$

Ultimately, eqs.(3-6),(3-2) and (3-5) yield

$$\mathcal{A}^{(\nu)}_{n,LM} = E_o\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu} \sum_{\bar{L}\bar{M}} \sum_{M'M''} \bar{U}^{(A)}_{n,LM';\bar{L}\bar{M}} W_{n,\bar{L}M''} D^{(L)}_{MM'}(\theta_\nu)\ D^{(\bar{L})*}_{M''\bar{M}}(\theta_\nu) \tag{3-7a}$$

and

$$\mathcal{B}^{(\nu)}_{n,LM} = E_o\, e^{i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu} \sum_{\bar{L}\bar{M}} \sum_{M'M''} \bar{U}^{(B)}_{n,LM';\bar{L}\bar{M}} W_{n,\bar{L}M''} D^{(L)}_{MM'}(\theta_\nu)\ D^{(\bar{L})*}_{M''\bar{M}}(\theta_\nu) \quad . \tag{3-7b}$$

Eqs.(3-7) evidence that the multipolar amplitudes of the scattered field, when referred to $\Sigma^{(\nu)}$, depend on the orientation of the cluster only through the elements of the matrices $\underset{\approx}{D}^{(L)}(\theta_\nu)$. The other quantities in eqs.(3-7), with the exception of the phase factor, do not depend on ν and can thus be calculated once for all. This result yields great advantage for the calculation of the optical constants of the medium. Indeed, the results obtained thus far, eqs.(3-7), can be related to macroscopic optics in a way that, although elementary, will maintain fairly visible the effects induced by the lack of spherical symmetry of the scattering centers.

The normalized forward-scattering amplitude of the ν-th cluster is [2]

$$\underset{\sim}{f}_{\nu,n} = \frac{e^{-i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu}}{E_o\sqrt{2}}\frac{1}{ik}\sum_{LM}(-i)^L[\mathcal{A}^{(\nu)}_{n,LM}\underset{\sim}{X}_{LM}(\hat{k}) + i\,\mathcal{B}^{(\nu)}_{n,LM}\hat{k}\times\underset{\sim}{X}_{LM}(\hat{k})] \quad . \tag{3-8}$$

Defining $f_{\nu,n'n} = \underset{\sim}{u}^*_{n'}\cdot\underset{\sim}{f}_{\nu,n}$, where $\underset{\sim}{u}^*_{n'} = \dfrac{\underset{\sim}{\varepsilon}_1 - in'\underset{\sim}{\varepsilon}_2}{\sqrt{2}}$ is the (complex) unit polarization vector, eqs.(3-8) and (2-2) yield

$$f_{\nu,n'n} = \frac{1}{8\pi ikE_o}\; e^{-i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu}\sum_{LM}W^*_{n',LM}(\mathcal{A}^{(\nu)}_{n,LM} + n'\mathcal{B}^{(\nu)}_{n,LM}) \quad . \tag{3-9}$$

These quantities are in turn related to the (complex) refractive index of the medium by [1]

$$\eta_{n'n} = \delta_{n'n} + \frac{2\pi}{k^2V}\sum_\nu f_{\nu,n'n} \quad , \tag{3-10}$$

where the sum includes the clusters within the volume V. We recall that $n_n = \mathcal{R}e(\eta_{nn})$ and $\gamma_n = 2k\mathcal{I}m(\eta_{nn})$ are the macroscopic refractive index and the absorption coefficient of the medium, respectively; the off-diagonal elements, $\eta_{nn'}$, account for the rotation of the plane of polarization. Since $f_{\nu,n'n} = f_{\nu',n'n}$ when the identical clusters ν and ν' have also the same orientation, eq.(3-10) can be rewritten as

$$\eta_{n'n} = \delta_{n'n} + \frac{2\pi}{k^2}\sum_i N_i\; f_{i,n'n} \quad , \tag{3-11}$$

where N_i is the number density of clusters having the i-th orientation. Provided the low density of the medium is not too low, eq.(3-11) can be replaced by

$$\eta_{n'n} = \delta_{n'n} + \frac{2\pi}{k^2} \int N(\theta)\, f_{n'n}(\theta)\, d\theta \quad . \tag{3-12}$$

In turn eq.(3-12) with eq.(3-9) give

$$\eta_{n'n} = \delta_{n'n} + \frac{1}{4k^3 i} \sum_{LM} W^*_{n',LM} (\bar{A}_{n,LM} + n'\bar{B}_{n,LM}) \quad ,$$

where

$$\bar{A}_{n,LM} = \frac{1}{E_o} \int e^{-i\underset{\sim}{k}\cdot\underset{\sim}{R}_\theta}\, \mathcal{A}_{n,LM}(\theta)\, N(\theta)\, d\theta \quad , \tag{3-13a}$$

$$\bar{B}_{n,LM} = \frac{1}{E_o} \int e^{-i\underset{\sim}{k}\cdot\underset{\sim}{R}_\theta}\, \mathcal{B}_{n,LM}(\theta)\, N(\theta)\, d\theta \quad . \tag{3-13b}$$

In eqs.(3-13) $\mathcal{A}_{n,LM}(\theta)$ and $\mathcal{B}_{n,LM}(\theta)$ refer to a cluster whathever of orientation θ and the phase factor $e^{-i\underset{\sim}{k}\cdot\underset{\sim}{R}_\theta}$ is appropriate to that cluster.

Eqs.(3-7) show that the integrations in eqs.(3-13) require only the calculation of the integrals

$$d(\bar{L}\bar{M}\bar{M}';LM'M) = \int N(\theta)\, D^{(\bar{L})*}_{\bar{M}'\bar{M}}(\theta)\, D^{(L)}_{MM'}(\theta)\, d\theta \quad . \tag{3-14}$$

This results in a further substantial simplification in the case that also the orientation of the clusters is random. In this case $N(\theta) = \frac{N}{8\pi^2}$, where N is the number density of all the clusters, and we have

$$d(\bar{L}\bar{M}\bar{M}';LM'M) = N\frac{1}{2L+1}\delta_{\bar{L}L}\delta_{\bar{M}M'}\delta_{\bar{M}'M}, \qquad (3\text{-}15)$$

on account of the orthogonality properties of the rotation matrices $\underset{\approx}{D}^{(L)}$(11). Then eqs.(3-13) with eqs.(3-7) and (3-15) become

$$\bar{A}_{n,LM} = \frac{N}{2L+1}\sum_{M'}\bar{U}^{(A)}_{n,LM';LM'}W_{n,LM},$$

$$\bar{B}_{n,LM} = \frac{N}{2L+1}\sum_{M'}\bar{U}^{(B)}_{n,LM';LM'}W_{n,LM}.$$

The limiting case when all the clusters have the same orientation is trivial. Assuming $\bar{\Sigma}^{(\nu)} \equiv \Sigma^{(\nu)}$, whe have $N(\theta) = N\delta(\theta)$ and eq.(3-14) give

$$d(\bar{L}\bar{M}\bar{M}';LM'M) = ND^{(\bar{L})*}_{\bar{M}'M}(0)D^{(L)}_{\bar{M}M'}(0) = N\delta_{\bar{M}'M}\delta_{\bar{M}M'}. \qquad (3\text{-}16)$$

Using eqs.(3-13), (3-7),(3-16) and (3-2), we find

$$\bar{A}_{n,LM} = \frac{N}{E_o} e^{-i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu} \mathcal{A}^{(\nu)}_{n,LM} ,$$

$$\bar{B}_{n,LM} = \frac{N}{E_o} e^{-i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu} \mathcal{B}^{(\nu)}_{n,LM} ,$$

where $e^{-i\underset{\sim}{k}\cdot\underset{\sim}{R}_\nu}$ cancels out with the phase factor contained in $\mathcal{A}^{(\nu)}_{n,LM}$ and $\mathcal{B}^{(\nu)}_{n,LM}$.

4. RESULTS

We applied the theory developed in the preceding sections to a few model media composed of clusters of rather simple structure with random space and orientational distribution. Each medium has been considered also for like orientation of clusters with several directions of $\underset{\sim}{k}$, the wavevector of the incident field. This allows us to show the effect of the orientational disorder of the scatterers on the optical constants of the medium. Table I reports the parameters defining the clusters of each medium individuated by a conventional name to which we will refer afterwards. Of course the names are descriptive; so "Dichro" denotes a simple medium capable to show an evident birefringence and dichroism. The coordinates of the centers and the radii of the spheres as well as $1/k$ are measured in arbitrary units. The radius of one of the spheres

has been labelled as b and chosen to enter the parameter $x=kb$ versus which all the results will be reported. Though x could clearly be related to the ratio between the size of the cluster and the wavelenght of the incident field, further information on this point can be gained by the parameter $\xi=kb_o$, where b_o, the radius of the smallest sphere containing just one cluster, is also reported in Table 1.

The results of our calculations are better evidenced introducing the quantities

$$\rho_\eta = \frac{n_\eta - 1}{n^{(o)} - 1} \quad , \quad g_\eta = \frac{\gamma_\eta}{\gamma^{(o)}}$$

where

$$\eta^{(o)}_{\eta\eta'} = (n^{(o)} + i\gamma^{(o)}) \delta_{\eta\eta'}$$

are the elements of the refractive index of the medium that one obtains by the Mie theory when the spheres in each cluster are considered as independent scatterers, i.e. disregarding the multiple scattering processes even among the spheres in the same cluster[7]. ρ_η and g_η are, in a sense, normalized quantities

and do not depend on any concentration of scatterers we choose in the low density range. All our calculations were performed for x ranging from 0.001 to 1.0 but all the results are actually reported for x between 0.1 and 1.0, the most interesting range as far as the effects of orientational order and disorder are concerned. It is worth noticing, in this connection, that when $x=1.0$, ξ reaches 2. for "Oxygen", 3.5 for "Water" and 3.1 for "Dichro". Therefore the wavelenght is always larger than the radius of the spheres but becomes comparable to the overall size of the clusters.

Figs. 1 and 2 report ρ_1 and g_1, respectively, for "Oxygen". The solid curves refer to a medium whose clusters are oriented alike for several values of the polar angle $\theta_{\underset{\sim}{k}}$, which, in this case, gives the angle between $\underset{\sim}{k}$ and the axes through the centers of the spheres. Quite identical curves could be drawn for ρ_{-1} and g_{-1}. As we expected the dependence of ρ_n and g_n on the orientation of the clusters increases with x[5]. The broken curves in figs.1 and 2 give ρ_1 and g_1, respectively, when the clusters are randomly oriented; they carry no label because for random orientation the results are independent of the direction of $\underset{\sim}{k}$. It is worth noticing the crossing of all the curves in fig.1, including the broken one,

at a single value of x. This result suggests that, as far as ρ_n is concerned, there exist a wavelenght at which the medium behaves as a cloud of spherical scatterers. This feature is shared in common with other media and may occur also for the curves of g_n.

Figs. 3 and 4 report ρ_1 and g_1, respectively, for "Water". Solid and broken lines have the same meaning as before and their behaviour suggests analogous considerations. The solid curves are labelled by the value of $\theta_{\underset{\sim}{k}}$, while $\phi_{\underset{\sim}{k}}=90°$. Of course the calculations were actually performed for several values of $\phi_{\underset{\sim}{k}}$. We also stress that the crossing of all the curves occurs both for ρ_1 and g_1 though at different values of x.

The last medium we consider in this paper is "Dichro" whose ρ_1 and g_1 are reported in figs.5 and 6, respectively. The complete lack of rotational symmetry of the clusters in this medium prevents all the curves to cross at a single point but all the crossings occur within a small range of x. The most striking features of this medium are its birefringence and dichroism which, though strongly attenuated, do not vanish even when the clusters are randomly oriented. We report in figs. 7 and 8 the quantities

$$\beta_n = \frac{n_n - n_{-n}}{n_n - 1} \qquad , \qquad d_n = \frac{\gamma_n - \gamma_{-n}}{\gamma_n}$$

respectively, for n=1; clearly $\beta_{-n} \simeq -\beta_n$ and $d_{-n} \simeq -d_n$. The differ-rent scale used in figs.7 and 8 for the case of random orientation should be noted.

The off-diagonal elements of $\mathcal{N}$, eq.(3-10), were also calculated for all the cases reported above and found to be non-vanishing but when expected according to the discussion of Newton(1). These results together with those reported above support the reliability of the theory developed in this paper to approximate the optical behaviour of a cloud of nonspherical model scatterers. We stress that our theory also applies when the orientational distribution of the clusters is not completely random. In this case it is necessary to estimate the d-integrals, eq.(3-14). In particular, if $N(\theta)$ is proportional to a generalized spherical harmonic (14) the d-integrals can still be given analitically. In any case, nothing prevents their use as disposable parameters, thus transforming the theory into a semiempirical method.

REFERENCES

1. R.G. Newton: "Scattering theory of waves and particles", (New York, 1966);
2. J.D.Jackson: "Classical electrodynamics", (New York, 1975);
3. M.Kerker: "The scattering of light", (New York, 1969);
4. M.Kerker, W.A.Farone and E.Matijevic: J.Opt.Soc.Am. 53,758(1963); W.A.Farone, M.Kerker and E.Matijevic: in "Electromagnetic scattering", ed. by M.Kerker,(Oxford, 1963); W.Heller: in "Electromagnetic scattering", ed. by M.Kerker,(Oxford, 1963);
5. R.T.Wang, J.M.Greenberg and D.W.Schuerman: Optics Lett. 6,543(1981);
6. J.M.Gerardy and M.Ausloos: Phys.Rev. B25,4204(1982);
7. F.Borghese, P.Denti, R.Saija, G.Toscano and O.I.Sindoni: Aerosol Res.Tech., to be published;
8. O.I.Sindoni, F.Borghese, P.Denti, R.Saija and G.Toscano: Aerosol Res.Tech., to be published;
9. F.Borghese, P.Denti, R.Saija, G.Toscano and O.I.Sindoni: J.Opt.Soc.Am., to be published;
10. V.K.Varadan, V.N.Bringi and V.V.Varadan: Phys.Rev. D 19, 2480 (1979);
11. M.E.Rose: "Elementary theory of angular momentum", (New York, 1957);
12. M.E.Rose: "Multipole fields", (New York, 1955);
13. F.Borghese, P.Denti, G.Toscano and O.I.Sindoni: J.Math. Phys. 21, 2754 (1980);
14. A.S.Davydov: "Quantum mechanics", (New York, 1965).

Riassunto

Viene presentato un metodo per il calcolo delle costanti ottiche macroscopiche di un mezzo di bassa densità costituito da una nube di diffusori identici non sferici. I diffusori del mezzo sono "cluster" di sfere dielettriche ed il campo elettromagnetico diffuso da ciascuno di essi è ottenuto come sovrapposizione di campi multipolari, come precedentemente proposto dagli autori. Facendo uso delle proprietà di trasformazione dei multipoli sferici sotto rotazione, i termini dipendenti dalla orientazione nell'espressione dell'ampiezza di diffusione in avanti vengono fattorizzati. In questo modo la somma delle ampiezze di diffusione di "cluster" con differenti orientazioni, necessaria per il calcolo della risposta ottica del mezzo, viene molto facilitata e assume una semplice espressione analitica quando i "cluster" sono orientati a caso. Vengono anche presentati i risultati del calcolo delle costanti ottiche per qualche mezzo modello.

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"Macroscopic optical constants..."

FIG.1

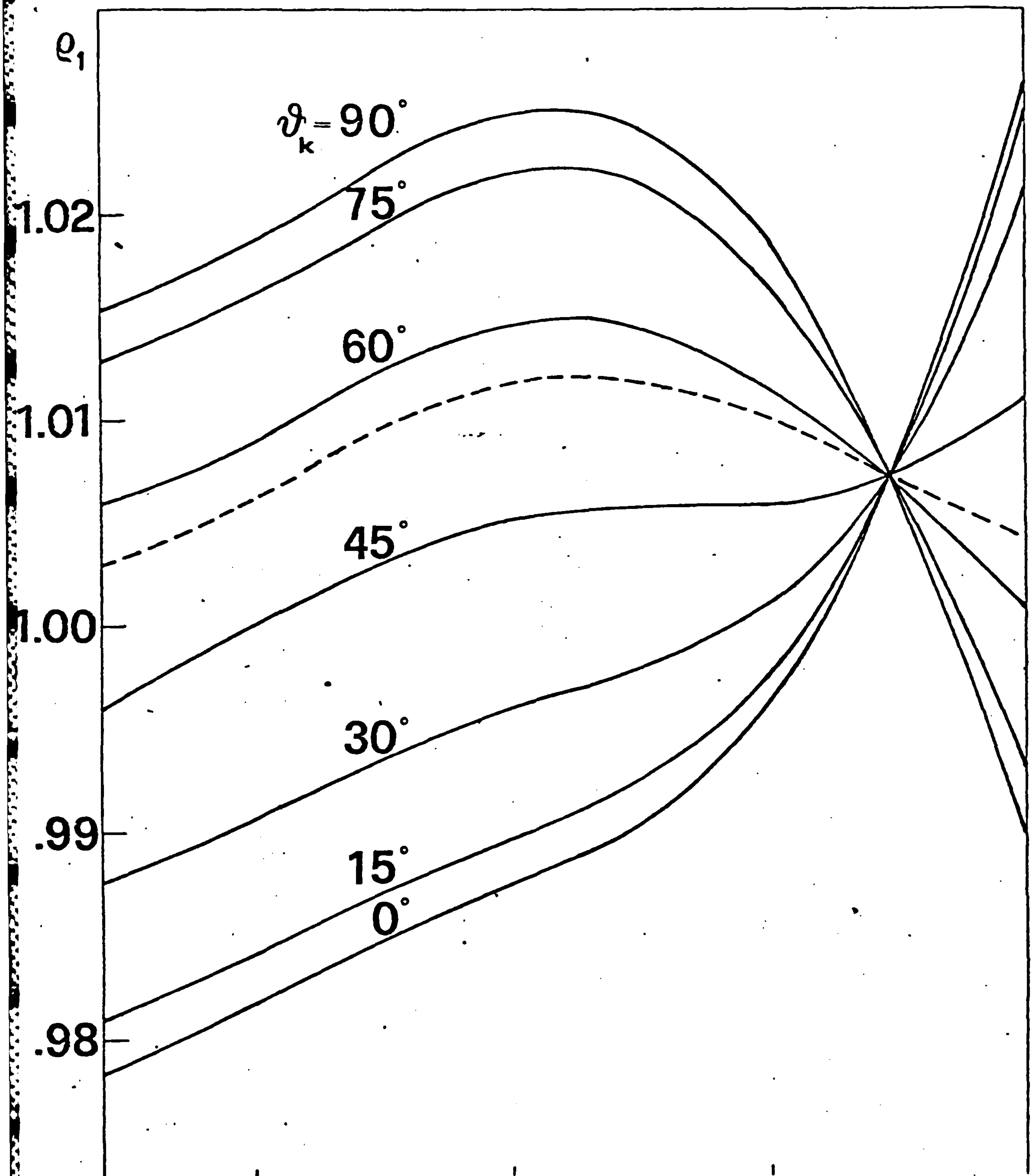

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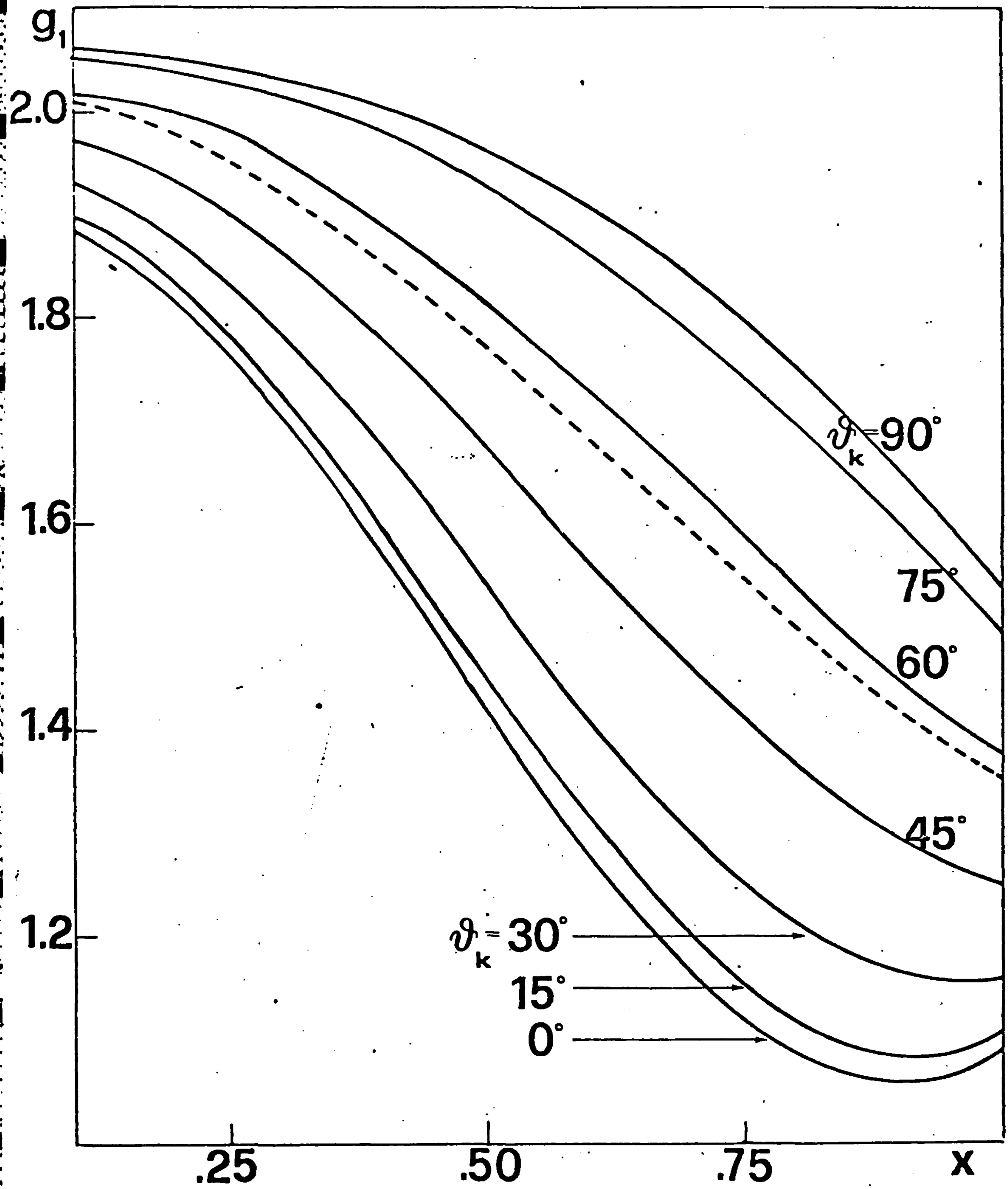
FIG.2



constants..."

FIG.3

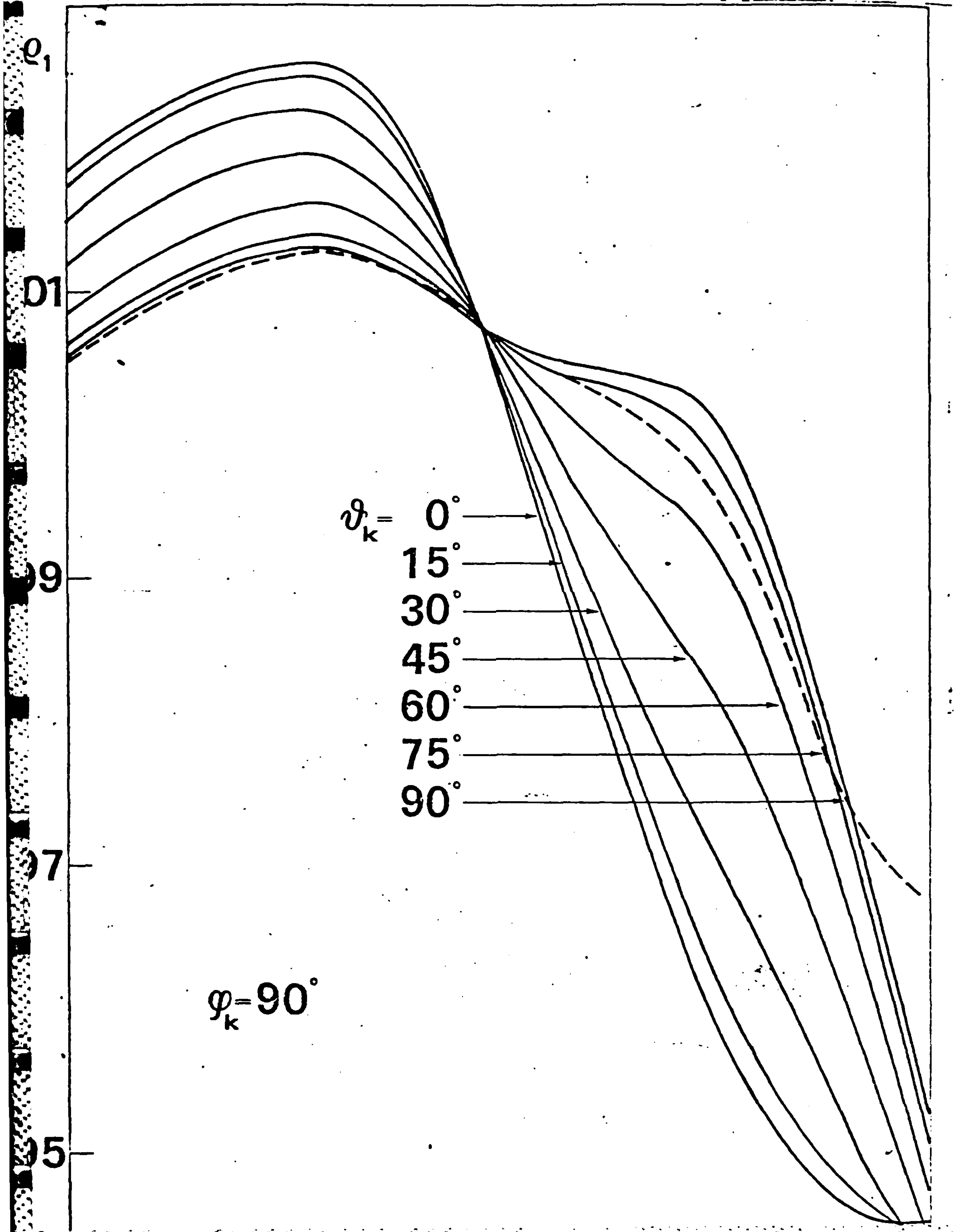


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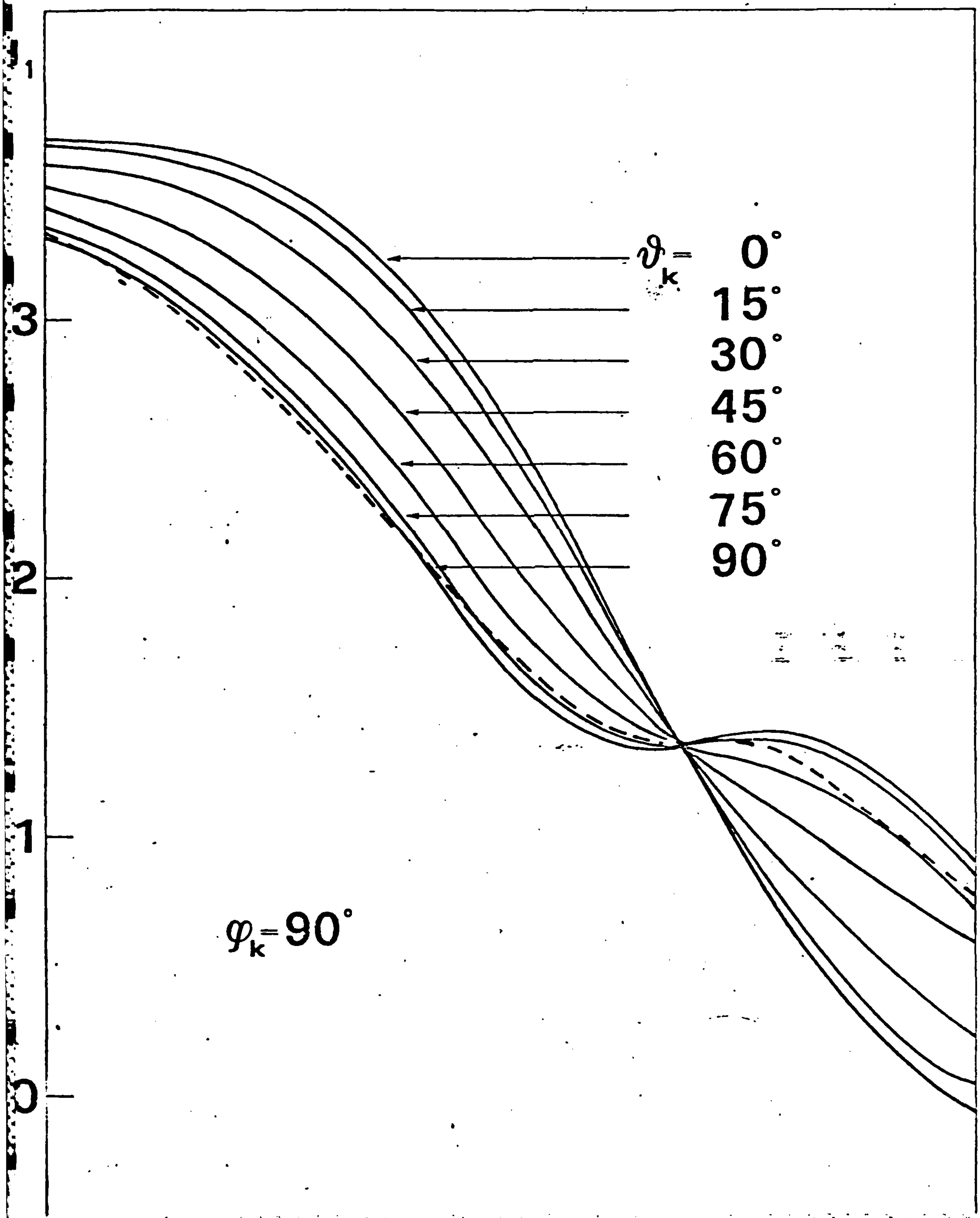
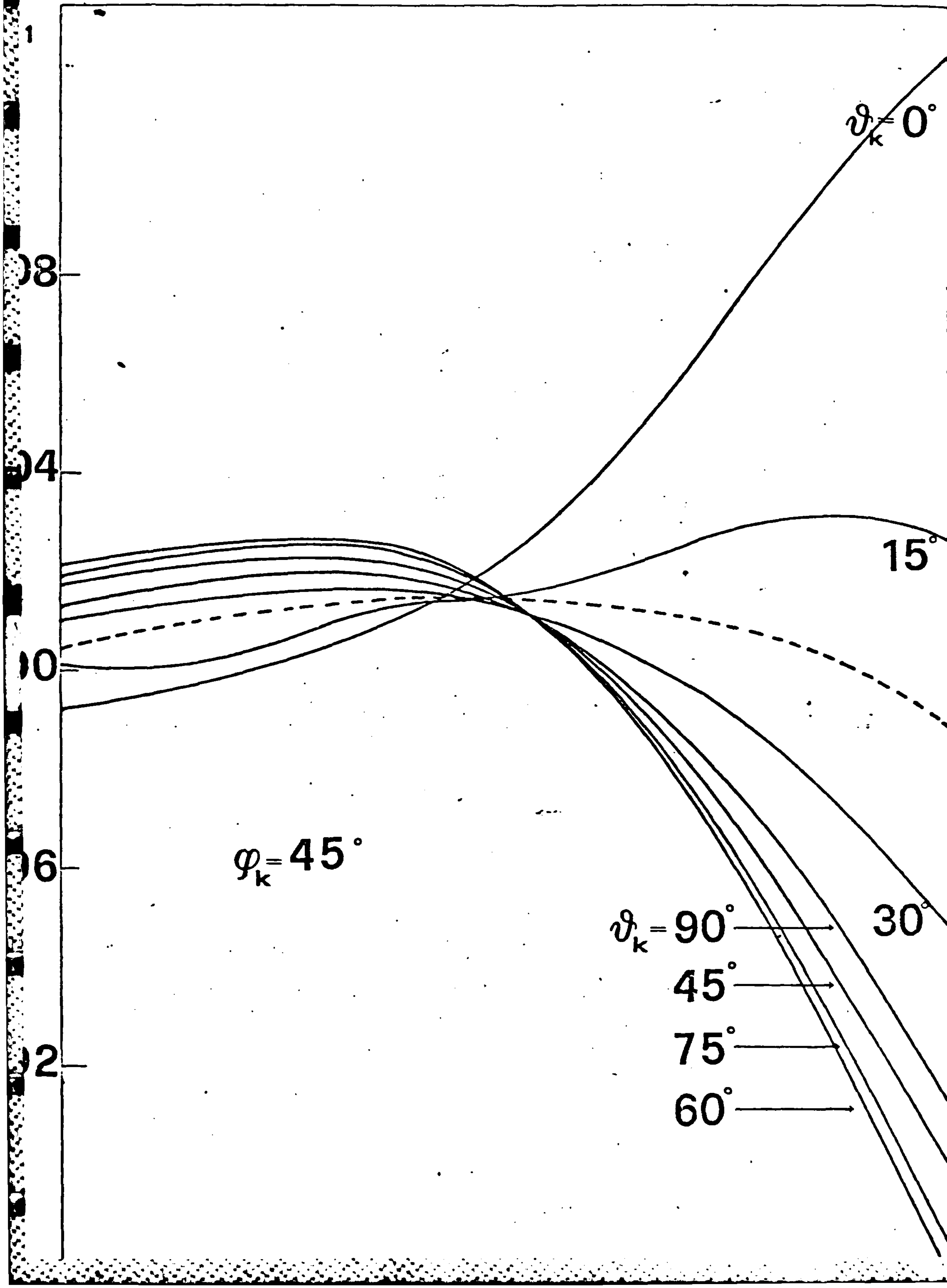
FIG. 4



constants..."

FIG.5



"...macroscopic optical constants..."

FIG. 6

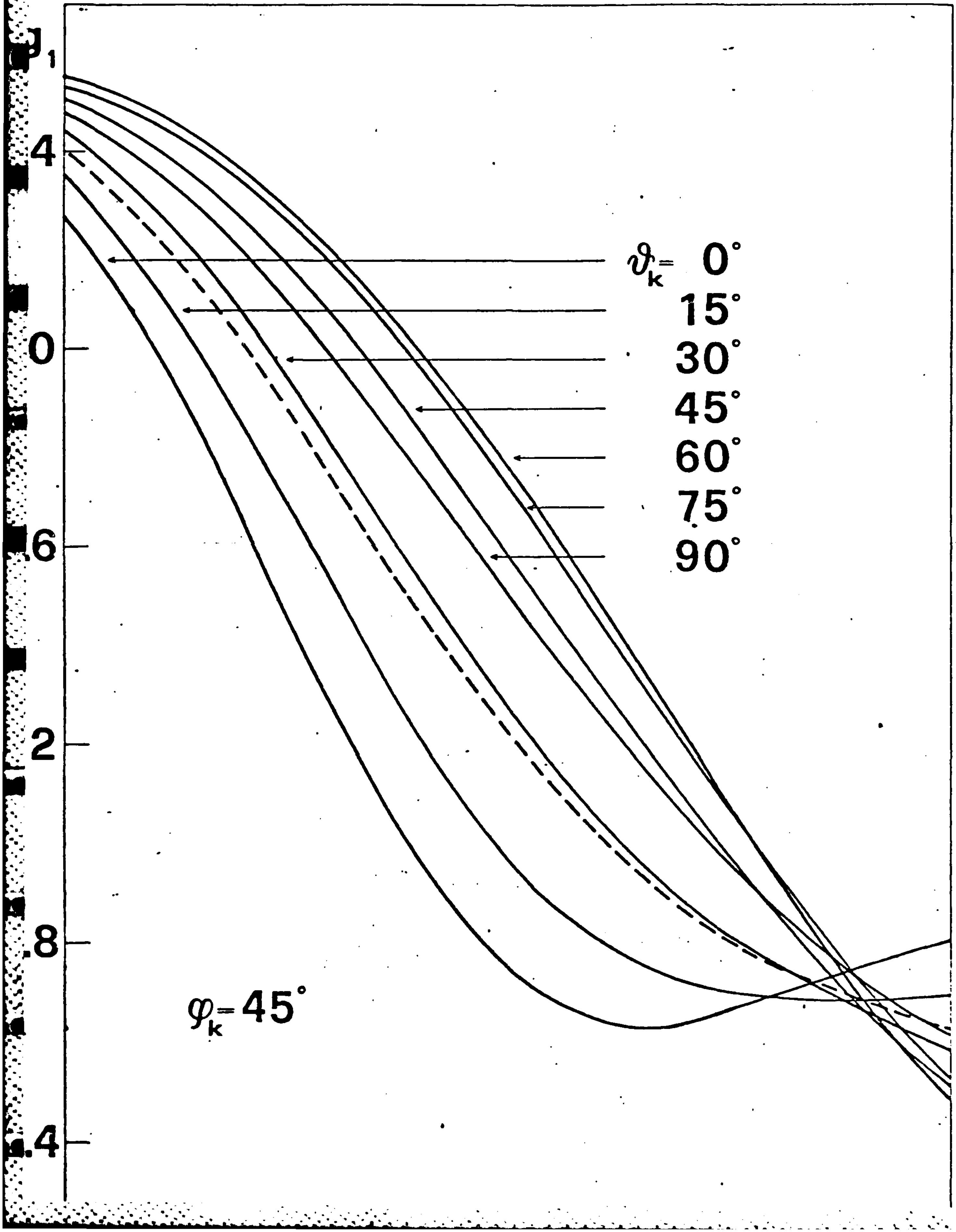
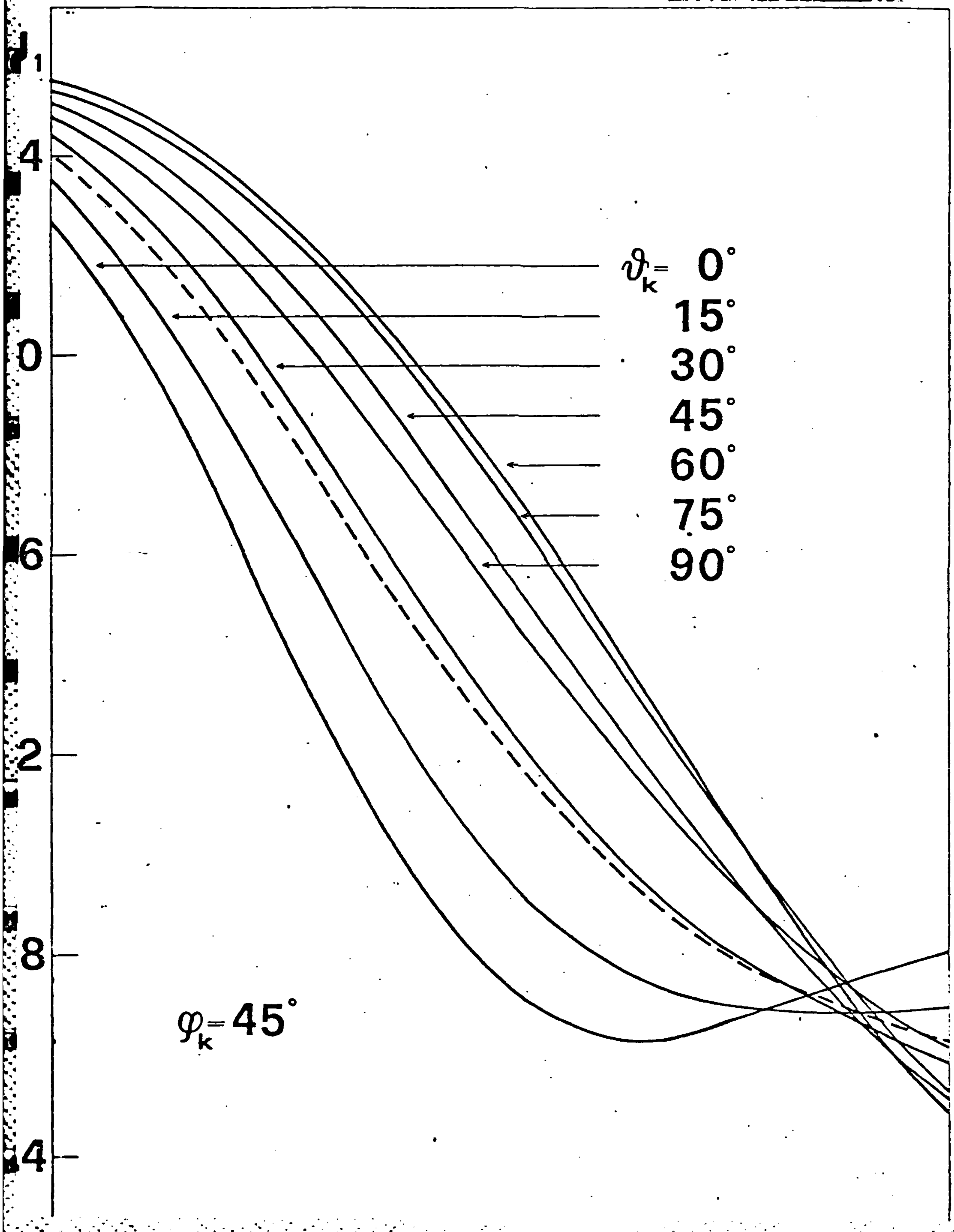
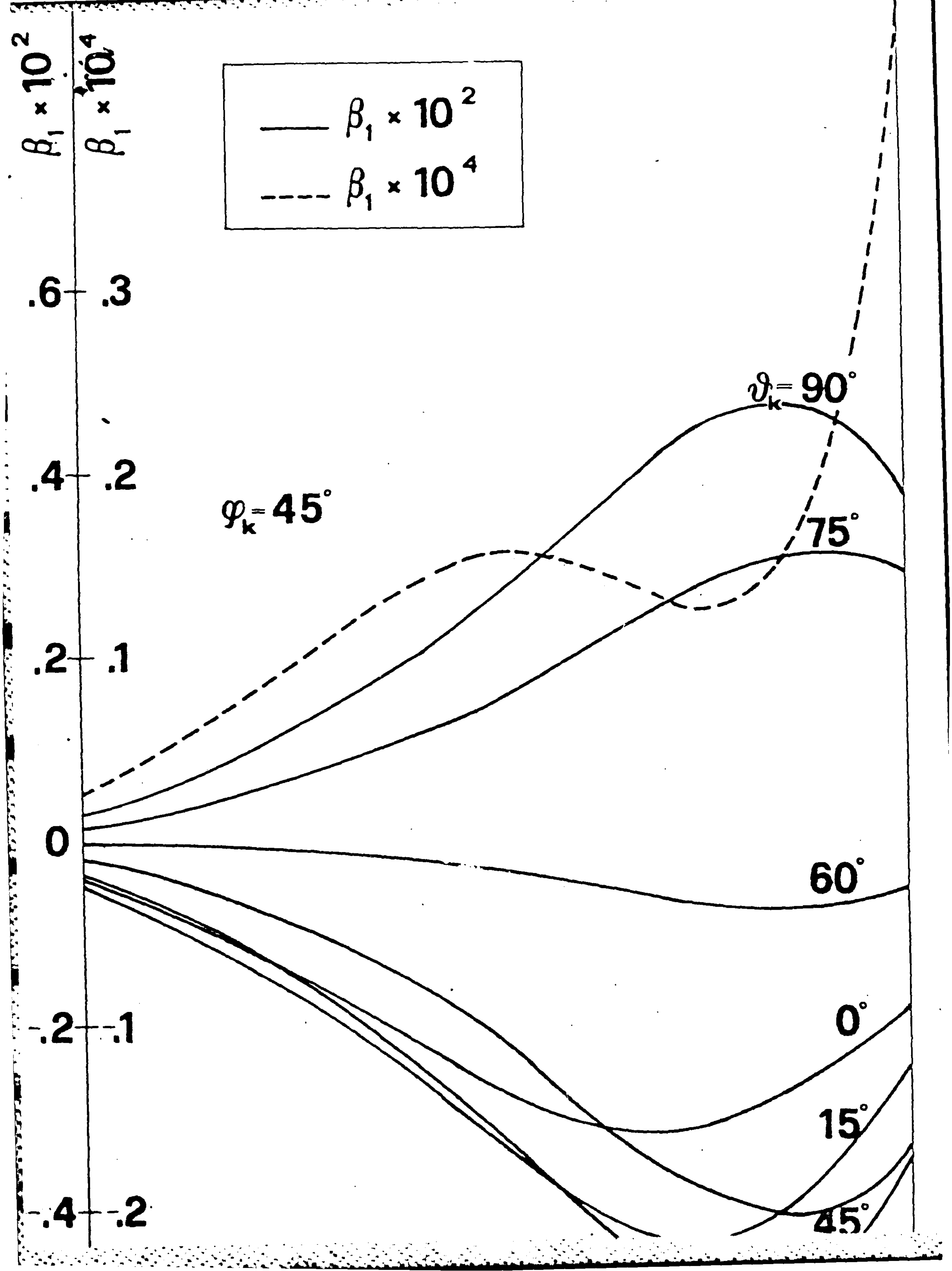
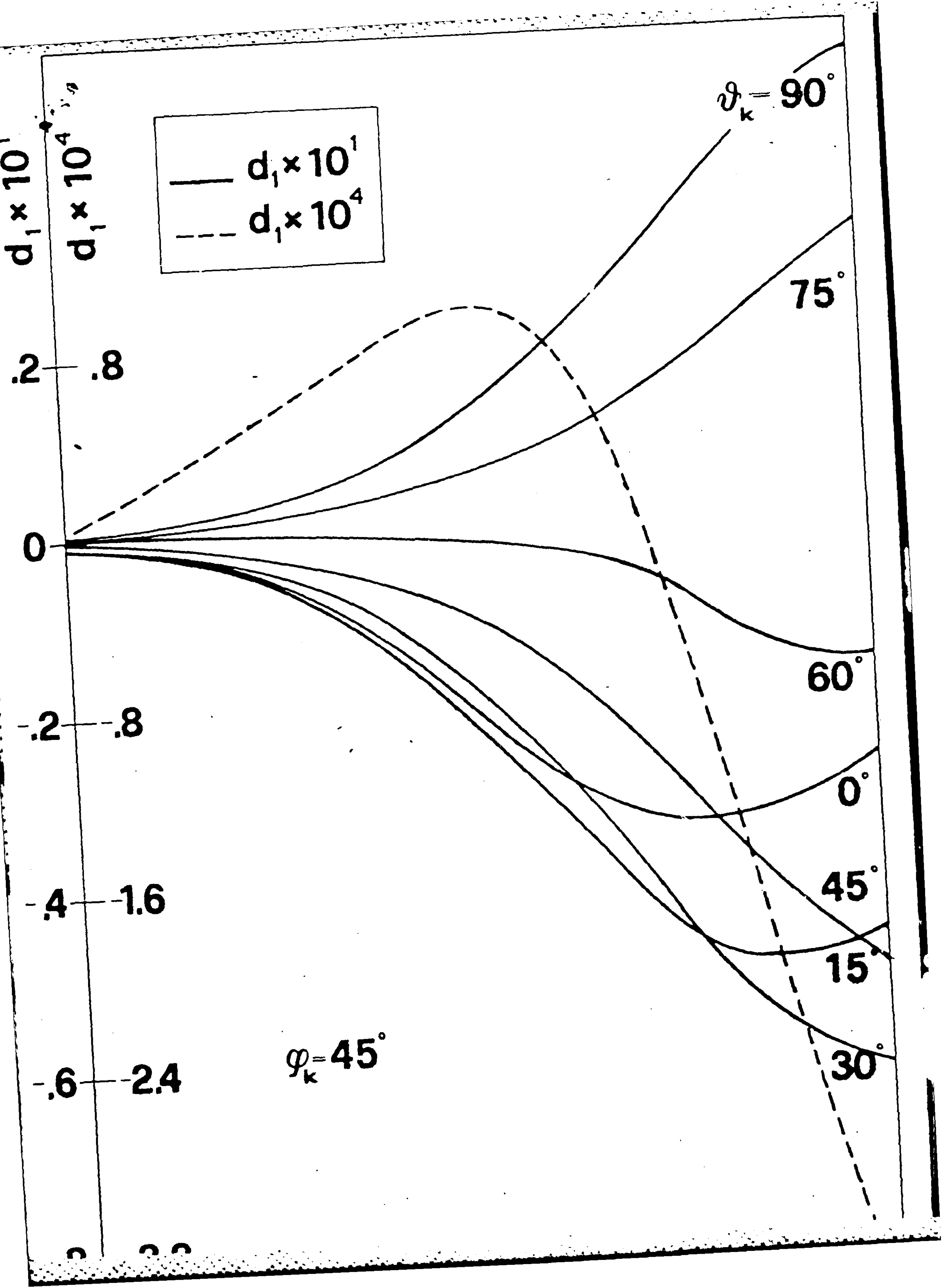


FIG. 6





$\beta_1 \times 10^2$
$\beta_1 \times 10^4$
$\beta_1 \times 10^2$
$\beta_1 \times 10^4$
.6 .3
.4 .2
.2 .1
0
-.2 -.1
-.4 -.2
$\varphi_k = 45^\circ$
$\vartheta_k = 90^\circ$
75°
60°
0°
15°
45°



$d_1 \times 10^1$
$d_1 \times 10^4$
$d_1 \times 10^1$
$d_1 \times 10^4$
$\vartheta_k = 90^\circ$
75°
60°
0°
45°
15°
30°
$\varphi_k = 45^\circ$
.2 .8
0
-.2 .8
-.4 1.6
-.6 2.4

FILM